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In a remarkably short time we have made tremendous progress in both the design and construction of a unique "atomic lithography" deposition facility, and in observing effects of an intense x-ray beam on the growth morphology of a thin semiconductor film. Now, with follow-on funding, we believe we are in a position to achieve the ambitious goals of this project. That is, to use a standing x-ray wave to template the growth of crystalline materials on amorphous substrates.

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Final Technical Report
AFOSR grant # F49620-97-1-0483, "Atomic Lithography"

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Introduction

During the previous grant period, we successfully conducted several experiments that demonstrate a direct effect of an intense x-ray beam on an atomic flux. In these experiments, we deposited Ge films in the thickness range 75 -225 Å onto amorphous substrates (amorphous Si oxide layers grown on Si wafers), both in the presence and absence of an x-ray beam. We characterized these samples using atomic force microscopy (AFM), transmission electron microscopy (TEM), and low angle x-ray diffraction (LAXRD). In brief, we see very clear differences between the morphologies of the in- and out-of-the-beam regions of the films, as well as find evidence for differences in the grain structure. We believe the evidence, which we present in more detail below, is strong that the effects we see are due to a direct interaction between the x-ray beam and the atomic flux. This is a very promising development indeed, and an important milestone towards producing monocrystalline films on arbitrary substrates by radiation-induced epitaxy. Below, we provide details on these experiments and the results.

Experimental Details

The x-ray beam generated by an 18 kW Cu-Kα rotating anode source is focused by a bent-crystal monochromator to an approximately 400 μm × 30 mm line on an ~5mm × 5mm amorphous substrate in our small UHV test chamber. This beam geometry allows us to produce both "beam-on" and "beam-off" samples simultaneously on the same wafer, thus completely eliminating sample-to-sample variations. It also provides large enough amounts of each sample to facilitate finding the beam-on sample, and to allow us to use several characterization methods. The x-ray beam is incident at 15° from normal, as illustrated in Fig. 1. Making the substrate smaller than the 30 mm length of the beam profile allows us to locate the beam photographically using fluorescent gadolinium oxysulfite painted onto special pads on the sample holder (not on the sample itself) and accurately index the beam-on area for measurement in the AFM. This is shown in Figure 2. The faint glow from x-ray beam in this photo can be seen traversing the 5 mm × 5 mm substrate (amorphous Si-oxide on a Si wafer) from the upper

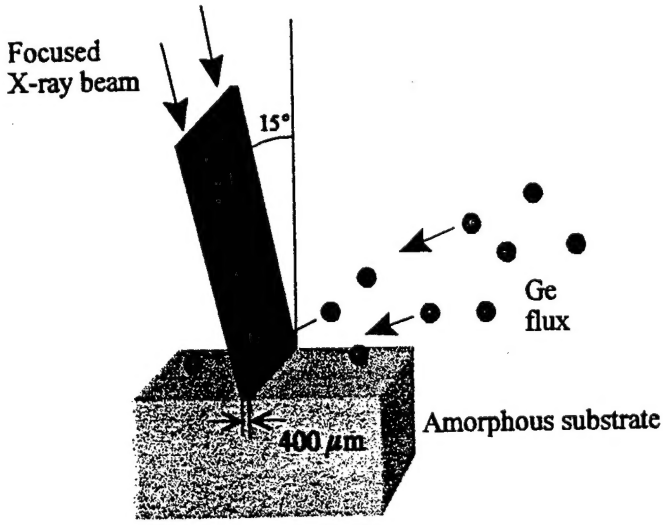

Figure 1 Experimental geometry for the Ge depositions described in this report.

right corner to the center of the lower side. The beam is made visible on either side of the wafer by the fluorescent coating on the sample holder, and the beam appears larger than the actual 400 μm width due to fluorescence caused by secondary electrons. The external light source illuminating the sample holder for this photo is not the Ge Knudsen cell, and therefore its direction does not indicate the Ge flux direction, which actually is from the lower right.

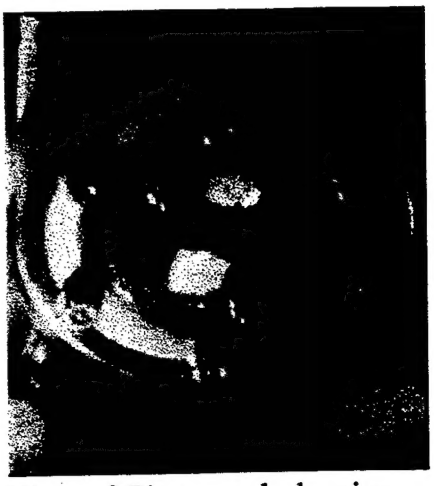

Figure 2 Photograph showing beam alignment in our small UHV test chamber.

The Ge flux is produced by a high-temperature Knudsen cell oriented such that the flux travels perpendicular to the x-ray beam at a 75° angle of incidence to the substrate (i.e. 15° from grazing, illustrated in Fig. 1). The deposition rates we used for these experiments was 7.5 Å/min, as calibrated by LAXRD. The substrate was not intentionally heated (nor cooled) during these depositions, and typically reached an equilibrium temperature of 150 °C due to the radiant heating from the Knudsen cell.

Comparison of Beam-On and Beam-Off samples

For the Ge that we deposited onto oxidized Si wafers outside the x-ray beam (i.e., what we term "beam-off" samples), we see a distinct and recognizable morphology. Possibly as a result of the shallow flux incidence angle, or due to residual artifacts on the Si wafer from the polishing process, these Ge surfaces exhibit anisotropic, elongated features as observed by AFM. These features, shown in Figures 2a and 2c below, are typically 200 Å wide and many thousands of Å

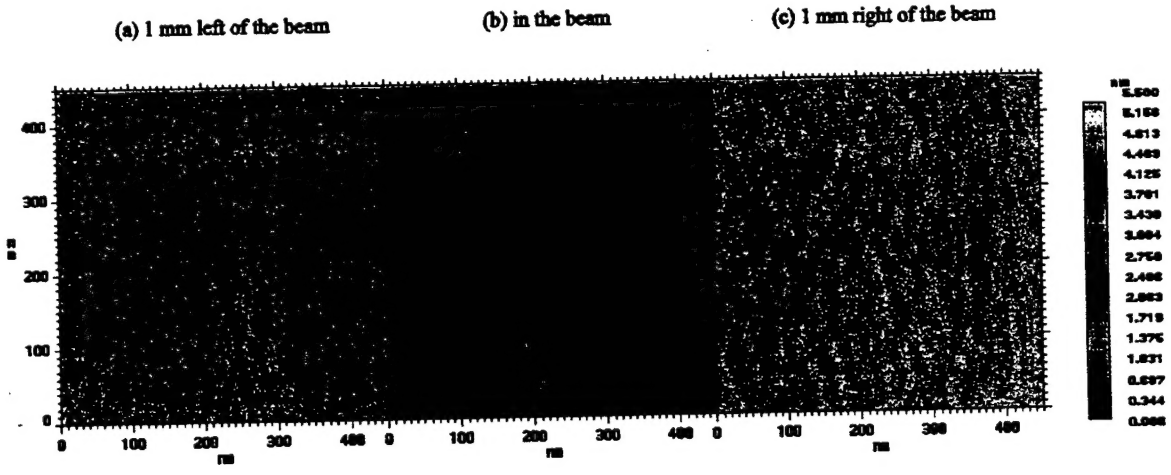

Figure 3 Comparison of beam-on (b) and beam-off (a,c) areas for a 75 Å Ge film grown on amorphous SiO_2. These images were taken from a single wafer, part of which was exposed to the x-ray beam during deposition. All three scans are plotted with the same vertical scale, indicating that the roughness of the beam-on area is significantly lower than that of the beam-off areas. As clearly seen, the morphology is also significantly different in the beam-on area.

long, with a peak-to-trough vertical distance of 30–60 Å The typical rms roughness of these surfaces is 8–12 Å for a 75 Å thick film. These features also are apparent in our planar TEM micrographs, which suggests that they may be more than just surface corrugation, but rather continue through the film's thickness. While the exact origin of these features is not known at this time, we note that they are always present in our beam-off AFM images, thus they provide a useful reference point for looking for effects of the x-ray beam.

In sharp contrast to the beam-off morphology, the Ge deposited in the portion of the substrate subjected to the focused 18 kW x-ray beam shows significant differences in both morphology and rms roughness. Figure 3 shows AFM images taken from three areas of one sample: the center from the beam-on region and the outer two from approximately 1mm to the left and to the right of the beam, respectively. The beam-on region, Figure 3b, is clearly much more isotropic than the beam-off regions, although slight remnants of the elongated features can be discerned. Also, the rms roughness in the beam-on region is a full factor of five smaller (or ~2–3 Å) than in the beam-off regions. In other words, the beam-on micrograph in Fig. 3b shows a *nearly atomically flat* surface over a 4500 × 4500 Å area.

The reduced roughness and different morphology strongly suggest that the growth dynamics are modified by the focused x-ray beam. Since the deposition rate is identical in the beam-on and beam-off regions, this implies a modification of the surface mobility of the Ge atoms. Furthermore, the similarity of the two scans on either side of the beam shows that the effect is in fact associated with the presence of the beam, and not merely variations in morphology at different points on the wafer. If further experiments unambiguously confirm that this effect is caused by direct momentum transfer from the radiation field inducing a change in surface mobility, it is very promising tool indeed for inducing a monocrystalline growth. However, we must first consider whether it could be due to some less useful effect, such as heating, photo-ionization, or photochemical modification of the surface prior to deposition. We discuss these possibilities next.

Potential Spurious Effects

A. X-Ray Beam Heating

Since surface mobilities are always temperature dependent, the possibility of surface heating due to the x-ray power should be considered. However, we showed in a previous report earlier this summer the heating power is at most 70 μW/cm^2, resulting in a temperature increase of only 1.4 μK. Although this estimate, based on the radiation density at the wafer and the thermal conductivity of the wafer, was somewhat simplified, it shows x-ray beam heating is a minuscule effect.

B. Photo-Ionization

The possibility exists that the morphological effects we observe are due to ionization of Ge atoms by x-ray photons, which might increase the reaction rates between the film and the background gas, or by the presence of surface charges. However, we note that even at the relatively high

growth pressures available in our mini-MBE test chamber (~1 × 10^{-8} Torr), the monolayer formation rate is many times longer than our deposition rate (in this case, ~4 ML/min). Therefore, even in the worst-case scenario where the sticking coefficients of the background gasses were 0 with the beam off and 1 with the beam on, there is still not enough gas to contaminate a significant percentage of the surface before it is buried. For this reason, beam-induced contamination is very unlikely. Although more experiments are needed to completely rule out surface charging, we also consider this likely to be a small effect. We note that in the experimental geometry shown in Fig. 1 the Ge flux passes directly through the x-ray beam to get to one of the beam-off regions that we imaged by AFM. The Ge atoms that pass through the beam, if ionized, would not de-excite until they hit the substrate. Therefore, if the beam-on region were affected by charging, we might expect a similar effect here, although perhaps not as large due to the small amount of time these atoms spend in the beam. However, the lack of a difference between the left and right beam-off regions (i.e. Figs. 3a and 3c) is consistent with a negligible charging effect.

C. Photochemical Modification of the Amorphous Substrate

Of the potential spurious effects, this one seems to us to be the one of greatest concern. The substrate is, by necessity, exposed to the intense x-ray beam for up to 90 minutes prior to deposition during the sample alignment and photographic beam locating procedure, so the possibility exists that the beam may alter the surface chemistry or roughness. Such a modification could occur by photon-induced desorption of residual adsorbed gases, photochemical reactions between adsorbed gases and the substrate, or even radiation damage to the substrate itself. Any of these could have an effect on the roughness and morphology of any film subsequently deposited. To test this possibility, we made one sample by first exposing an amorphous SiO_2 substrate to the x-ray beam for the same length of time (~90 min) as the samples are normally exposed during alignment. Immediately following this pre-exposure we closed the x-ray shutter

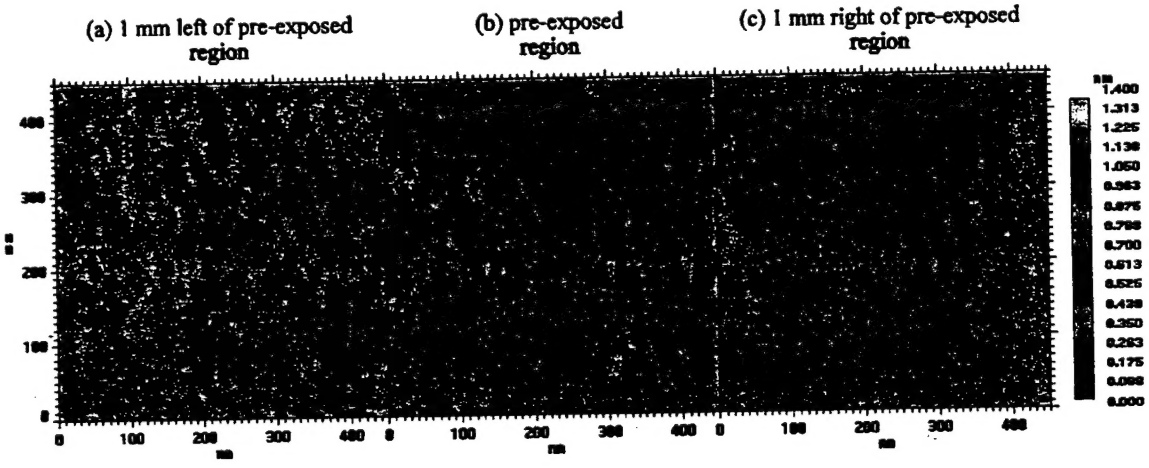

Figure 4 Comparison of Ge deposition on an amorphous SiO_2 substrate that had been pre-exposed to the x-ray beam for 90 minutes (b), and not exposed (a,c). After the pre-exposure, the beam was turned off, so that no beam was present during deposition. As in Fig. 3, the Ge thickness is 75 Å.

and deposited a Ge film of the same thickness as the one imaged in Fig. 3. However, in this case no x-ray beam was present during the actual growth. The results of AFM studies of this film, with images taken in the region where the pre-exposure beam had been incident, as well as on both sides of that region, are shown in Fig. 3. With the exception of small differences in the surface roughness, all three of the surfaces in Figure 4 are essentially identical. Significantly, there are not the dramatic differences in morphology that are shown in Fig. 3 with the sample grown with the x-ray shutter open. Furthermore, the morphology of all three portions of this test sample generally resembles the beam-off areas of Fig. 3. This is further evidence that the effect we observed is due to a direct interaction of the x-ray beam and atomic flux.

Proposed Future Work

The goal of this project, templated monocrystalline growth on amorphous substrates, is a very ambitious one. However, we believe the results briefly discussed in this report—observation of a direct effect on the crystal structure of a thin film due to an intense focused x-ray beam—represents significant advancement toward that goal. Here, we outline how we would propose to exploit these results, as well as the facilities we specifically designed and constructed for this work, if continued funding were made available.

The obvious next step is to investigate the crystalline nature of these films using an x-ray beam in our full-scale, heavily-instrumented, MBE with its *in situ* AFM/STM, RHEED, and Brillouin light scattering facilities. We designed and constructed this special-purpose MBE machine specifically for this project, and it is now installed on a vibration isolation platform in our clean room ready to interface with our 18 kW rotating anode x-ray source. Since our custom MBE system is now ready and, having demonstrated the direct effect of x-rays on the growth, we are now ready to use the improved vacuum and *in situ* characterization facilities to resolve some of the unanswered questions about the effects we have found, as well as begin the all-important templating experiments using an intense standing wave. With a small investment (relative to the original equipment purchase), this project is positioned to accomplish its goals and provide the basis for much future work. Specifically, we outline below what we would propose to do during a continuation of the program.

Bringing x-rays into our full-scale MBE system would be accomplished by incorporating an additional tube on our existing 18 kW power supply, then introducing the x-rays into the chamber with minimal loss through a UHV beryllium window. While our small MBE has been an excellent test bed over the past year, it is limited in the materials it can produce. Although Ge is physically interesting and a good test material, it is of limited practical application. However, our full MBE system is equipped with an electron beam evaporator as well as Knudsen cells, which will allow us to perform experiments on Si and other more technologically important materials not currently possible with the single Knudsen cell in the small MBE chamber.

The next significant milestone will be the unambiguous demonstration of an x-ray standing wave. The results described earlier in this report do not depend on a standing wave being present; only

momentum transfer to the Ge atoms. Simply because the small MBE does not have the ability to precisely align the sample, we have made no attempt yet to create a standing wave. However, it should be relatively easy at this point for us to create a standing wave by Bragg reflection from a suitable crystal or multilayer, although the intensity, coherence and spatial extent of the wave must be characterized before we attempt to use it for radiation-induced epitaxy. We propose to accomplish this by luminescence experiments on thin films of a "marker" material deposited directly above the x-ray mirror to map the intensity profile of the wave. To do this, we would first convert our small MBE to a photoluminescence chamber with a spatially-selective photomultiplier and a modified sample holder to allow for precise alignment. We would then use this arrangement to evaluate different crystals as standing wave generators while the initial deposition experiments are being completed in the large MBE. Once the appropriate geometry is determined and the characteristics of the standing wave mapped out, the standing wave optics can then be quickly moved to the large MBE system, which is already equipped with the necessary high-precision five axis sample positioner.

Finally, by next summer these two parallel efforts will be brought together in the full-scale MBE system with the growth of the first of several materials (including Si) under the influence of the standing wave, and characterized of the samples by the extensive *in situ* facilities we specifically included when we designed this system. Also, during this time we will experiment with different wavelengths and detuning parameters to optimize the interaction of the x-ray beam with the atomic flux. We envision the growth of an amorphous/monocrystalline multilayer as the most effective demonstration that this effect has the potential for generating stacked integrated circuits that are isolated from each other, thus allowing greatly increased effective device densities.

Summary

We have observed significant effects on the growth of Ge onto amorphous SiO_2 from an intense x-ray beam which we believe demonstrate a coupling of the Ge flux to the radiation field. These effects include a dramatic reduction in the surface roughness and a modification of the natural surface morphology that Ge normally takes when grown on an amorphous substrate. While at this point we are not able to verify the crystallinity of the beam-on sample due to the limitations of our small test chamber (limitations that do not exist with our now-completed full MBE system), we believe this development is very promising for the possibility of radiation-induced templating. The results presented here were limited by the lack of RHEED in our test chamber and the fact that AFM had to be done *ex situ*. However, we are now ready to move these experiments to our full-scale MBE system equipped with the full range of required *in situ* structural characterization and deposition facilities. We believe these instruments would allow us to resolve the existing unanswered questions regarding the Ge deposition. More important, we would then be able to proceed with the full atomic lithography experiments we planned from the start.

In brief, in a remarkably short time we have made tremendous progress in both the design and construction of a unique "atomic lithography" deposition facility, and in observing effects of an intense x-ray beam on the growth morphology of a thin semiconductor film. Now, with follow-on

funding, we believe we are in a position to achieve the ambitious goals of this project. That is, to use a standing x-ray wave to template the growth of crystalline materials on amorphous substrates.